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The Titan haze simulation experiment: Investigating Titan's low-temperature atmospheric chemistry in a pulsed plasma jet expansion

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Abstract. The Titan Haze Simulation (THS) experiment is a unique experimental platform that allows to simulate Titan's complex atmospheric chemistry at low Titan-like temperature by generating a plasma discharge in the stream of a plasma jet expansion. Both gas and solid phase products are generated and can be analyzed using different in-situ and ex-situ diagnostics. Here, we present an overall description of the work accomplished with the THS in the last 10 years, our current research efforts, and the important implications for the analysis of Cassini's returned data and preparation for future Titan missions.

Keywords. planets and satellites: Titan, atmospheric effects, methods: laboratory

1. Introduction

In Titan's atmosphere, a complex organic chemistry induced by UV radiation and electron bombardment occurs at low temperatures (<200 K) between its main constituents, N_2 and CH_4, and leads to the production of heavy molecules and subsequently solid aerosols that form the haze layers that have been observed by Voyager and Cassini. Because the reactive carbon and nitrogen species present in Titan's aerosols could meet the functionality requirements for precursors to prebiotics, the study of Titan's aerosol has become a topic of extensive research in the fields of astrobiology and astrochemistry. Many experiments have been developed in order to understand Titan's gas phase chemistry and the production processes and composition of the atmospheric organic aerosols.

2. The Titan Haze Simulation (THS) experiment

The THS experimental setup was developed at the NASA Ames Cosmic simulation (COSmIC) facility. The unique characteristic of THS is that it uses a pulsed discharge nozzle (PDN) to generate a supersonic expansion that cools down a gas mixture to Titan-like temperature (150 K) *before* inducing the chemistry by plasma discharge (Biennier *et al.* 2006). In addition, because of the accelerated gas flow in the expansion, the residence time of the gas in the active plasma region is less than 4 microseconds. This results in a truncated chemistry that enables us to control how far in the chain of chemical reactions the chemistry is allowed to process. By adding, in the initial N_2-CH_4 gas mixture, heavier molecules that have been detected as trace elements on Titan, we can then study the first and intermediate steps of Titan's atmospheric chemistry as well as specific chemical pathways. Finally, because THS uses a jet expansion, the gas and solid phase products of the plasma-generated chemistry are accelerated to supersonic speeds before being detected or deposited for future analyses, which is the closest laboratory simulation of a probe descent in Titan's atmosphere. Below is a summary of the gas phase and solid phase studies that have been conducted with the THS in recent years.

Gas phase analyses: In the THS, a reflectron Time-of-Flight mass spectrometer (TOF-MS) is coupled to the PDN to monitor *in situ* the products of the chemistry (Ricketts *et al.* 2011). By using polarized skimmers that attract the plasma-generated ions present in the expansion, we can directly detect the ions in the TOF-MS without fragmenting them. In a mass spectrometry study of the gas phase, the unique capability of the THS to probe the first and intermediate steps of the N_2-CH_4 chemistry was demonstrated (Sciamma-O'Brien *et al.* 2014). This finding was further confirmed by a subsequent modelling study of the THS system using a one-dimensional model to simulate and track the evolution of more than 120 species in the THS (Raymond *et al.* 2018). In these studies, acetylene, ethylene and benzene were added to the initial N_2-CH_4 mixture and both the experimental data and the model showed more complex chemical growth when heavier precursors were present. The results of the experimental gas phase study have been compared to observational data from the Cassini Plasma Spectrometer Ion Beam Spectrometer (CAPS-IBS) and have provided very encouraging results that illustrate the unique power of the THS laboratory approach to help analyze and better understand returned data from Cassini's instruments. Efforts are now underway to modify the experimental setup and accommodate the detection of negative ions with TOF-MS in the THS to be compared to the negative mode data from CAPS.

Solid phase analyses: The THS can also be used to produce Titan aerosol analogs (also called tholins). THS tholins produced in the same gas mixtures as for the gas phase analysis were analyzed ex-situ using Scanning Electron Microscopy (SEM), X-Ray Absorption Spectroscopy (XANES), mass spectrometry, and visible and infrared (IR) spectroscopy, in order to investigate the effect of the initial gas mixture on their morphology and composition. SEM images confirmed the findings of the mass spectrometry: grains produced in more complex mixtures are much larger than those produced in N_2-CH_4 mixtures, consistent with a more complex chemistry occurring when adding heavier precursors. Mid-infrared spectroscopic analyses have highlighted a change in the nitrogen chemistry and an increase in aromatic abundance when benzene is present. A comparison of THS mid-IR spectra to Cassini Visible Infrared Mapping Spectrometer (VIMS) data has shown that the THS aerosols produced in simpler mixtures i.e., samples that contain more nitrogen and incorporate this nitrogen in isocyanide-type molecules instead of nitriles, are more representative of Titan's aerosols (Sciamma-O'Brien *et al.* 2017). Ongoing studies are investigating the optical properties of THS tholins to determine their optical constants from the visible to the far infrared, for the interpretation of observational data.

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